8273-EN-01
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The optical properties of aerosols

Final Technical Report
by
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August 1998

United States Army
EUROPEAN RESEARCH OFFICE OF THE U. S. ARMY
London, England

Contract number N68171-97-M-5546
Contractor: Centro Siciliano per le Ricerche Atmosferiche
e di Fisisca dell'Ambiente

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The research performed under the present contract has been addressed to the following topics;
1. Optical properties of aggregated spheres deposited on a dielectric surface with a random distribution of their orientations.
2. Determination of atmospheric pollution through spectral analysis of starlight.
The reserach mentioned above has been reported in a number of papers that were already printed or are at present in the process of printing.
At the same time, our reserach called our attentiuon on a number of mathematical problems thatn if not properly façed, may frustrate any attempt to get reliable numerical resuts.

The main findings that are reported in our papers will now be briefly summarized, and the lines of future progress are sketched.

1. Optical properties of aggregated spheres deposited on a dielectric surface with a random distribution of their orientations.
This research is meant to extend to the case of aggregated spheres the theory that we already developed to deal with the extinction spectra from spheres deposited on a dielectric substrate. The need for such an extension is easily understood on account that spherical particles tend to aggregate to form compound scatterers whose otical properties are known to be rather different from the the properties of the component spheres. Of course, the motivation of our study is to supply a reliable method to check the cleannes of surfaces.
Our starting point has been the theory that we already developed to deal with the spectra from aggregates of known orientation. Using the transformation properties of the spherical multipole fields under rotation we were able to define the transition matrix for an aggregate in the presence of a dielectric substrate and to perform analitically the required averages over the orientational distribution. As a result we were able to produce the full scattering patterns from a dispersion of binary aggregates deposited on a dielectri substrate with a random distribution of their orientations, including both co-polarized as well as cross-polarized patterns. The interpretation of our results has been reported in a paper that, though not yet printed, has been accepted for publication in JOSA A.
An important point regards the comparison of the results of our theory with the experimental data. To this end, the experimental group at the Dipartimento di Fisica della Materia e Tecnologie Fisiche Avanzate, was able to devise an experimental setup to record the spectra from a surface sparsely seeded by particles. Wen the resulting recoreded spectra were compared with the results of our calculations, a complete agreement was found within the experimental error.

2. Determination of atmospheric pollution through spectral analysis of starlight.

It is well known that the optical methods are a useful tool for the determination of atmospheric pollution. As a rule the light that serves as a probe is generated on the surface of Earth as in LIDAR techniques, or comes from satellites in appropriate orbits. In recent times our attention has been drawn on a third possibility, i. e. on the spectral analisis of the light that comes from selected stars. In principle, the proposed tecnique is rather simple. The light from a star is taken form two observers located at a different altitude is analized and the resulting spectra are compared. It is intuitive that any extra absorpion line that might appear in the spectra taken at low altitude should originate from atmospheric pollution. In practice the realization of this kind of measurements requires the presence in the same area, of an astronomical observatory and of a low altitude laboratory. A situation of this kind occurs in the area of Catania, Italy, where one can use the facilities of the Astronomical Observatory on Mount Etna, some 2900 metres of altitude, as well as the Laboratory of the University of Catania, that is located almost at sea level. Note that the horizontal distance between the Observatory and the Laboratory does not exceed 10 kilometres. Similar situations occur at other places in the World, e. g. in Chile, South America.
From a physical point of view, the actual determination of the polluting substances is complicated from the fact that, specially in the infrared, one should take into account the effects that are due to the presence of particles. The effect of the latter objects can be discarded from the observed spectra provided that one is able to calculate the extinction spectra from a polydispersion of particles. It is at this stage that the tecniques that we developed under the present and the previous Contracts enter into play. Indeed, even the most anisotropic particles can be dealt with by our codes, provided that the spectroscopic data on their chemical composition are known. At present, this researech is performed by dr Giovanni Catanzaro as a part of his work to get the degree of Dottore di Ricerca in Fisica. Of course, the work is far from concluded and we plan to pursue the subject in the framework of the next Cotract that has already been awarded by the ERO.

I cannot conclude this report withuot mentioning that thanks to the work performed under the present Contract, dr Maria Antonia Iatì, who collaborated to the work of the last two yiears, got the degree of Dottore di Ricerca in Fisica and is presently performing post doctoral research at the University of Waterloo, Ontario, Canada, under the direction of prof. Duley.

Pattern of the scattered intensity from binary clusters on a dielectric substrate

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Abstract

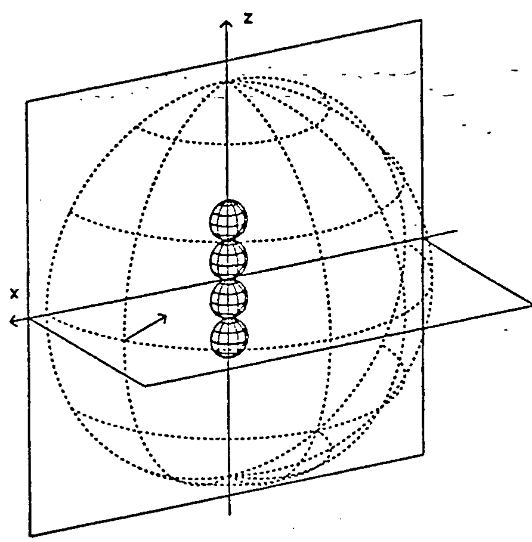
The preliminary results of the calculation of the full pattern of the scattered intensity from a dispersion of randomly oriented binary aggregates of identical spheres deposited on a dielectric substrate is presented.

In a recent paper we presented the full pattern of the scattered intensity from a single sphere deposited on the plane surface that separates two homogeneous media of different refractive index.[1] The theory behind our results is based on the expansion of the electromagnetic field into a series of spherical multipole fields and on the imposition of the boundary conditions across the plane surface and across the surface of the sphere. The difficulty of imposing the boundary conditions across the plane surface to a field given in terms of spherical multipole fields was overcome through the application of a general reflection rule for spherical vector multipole fields on a plane surface.[2] According to this rule, the reflection of a spherical multipole field with origin at P originates a linear combination of spherical multipole fields that satisfy the radiation condition at infinity, with origin at P', the mirror image of P with respect to the plane surface. Using this rule, that clarifies the role of image theory when the surface is not perfectly reflecting, we get a scattered field that has the correct behavior at infinity. Perhaps, the most distinctive feature of our approach is the fact that, unlike similar approaches to the same problem,[3,4] we get a non-vanishing field that propagates along the surface. As a result the patterns that we presented were in excellent agreement with the experimental findings and with the ab initio simulations.[5,6]

The purpose of this paper is to present some results for the full scattering pattern from binary aggregates of identical spheres deposited on a plane surface. The extension of our theory to the case of aggregated spheres is far from trivial because one has to consider the effect of the mutual interaction of the aggregated spheres: in other words, we had to modify our theory to account for the dependent-scattering effects in the presence of the plane surface.

Of course, aggregated spheres form anisotropic objects whose patterns are expected to have a noticeable dependence on their orientation with respect to the

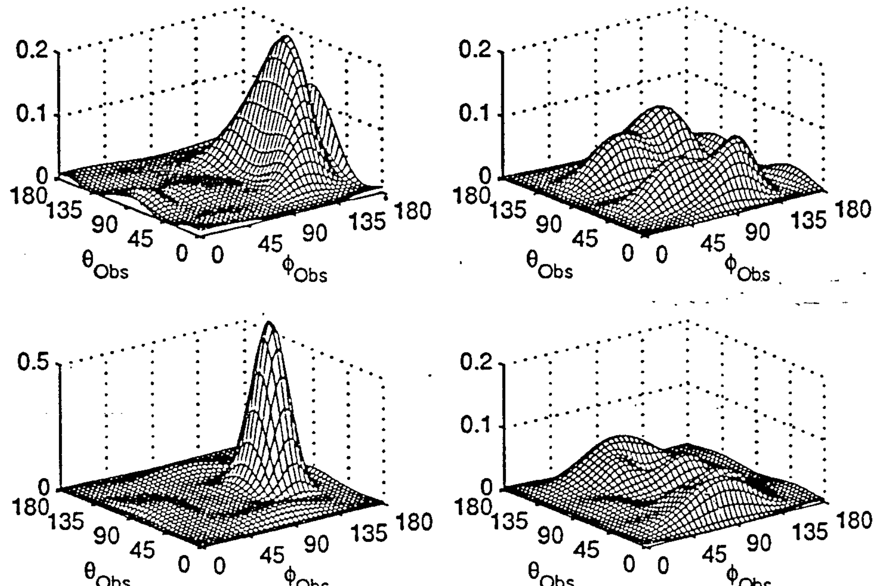
incident field. The field scattered by a dispersion of such objects even when deposited on a surface with a given distribution of their orientations may be rather different from the field scattered by a dispersion of the same objects all oriented alike. Nevertheless, by making full use of the transformation properties of the spherical multipole fields under rotation[7] we are able to calculate, through an analytical average, the field scattered, for instance, by an orientationally random dispersion of aggregates on the surface.[8] This has been achieved by giving a suitable definition of the transition matrix[1,9] of the aggregate in the presence of the surface. The elements of the transition matrix include, of course, all the interactions among the spheres that compose the aggregate. The resulting patterns, that will be discussed below, should give a reliable description of the physical situation that is likely to be meet in actual experiments.



In the figure above we sketch the geometry that we use to report our results for the scattering patterns. The surface of the substrate is the x-z plane and the direction of incidence is defined by the polar angles ϑ_I and φ_I. In particular for the results reported in this paper we chose $\vartheta_I = 90°$ and $\varphi_I = 225°$, i. e. the incident wavevector forms an angle of 45° with the normal to the surface. The polarization both of the incident and of the scattered field was chosen to lie either along the meridians (ϑ-polarization) or along the parallels (φ-polarization) of the large sphere sketched above.

In the upper part of the figure below we report, for purposes of comparison, the patterns generated by single spheres of radius $\rho = 450\,\text{nm}$ and refractive index $n = 2.0$ deposited on a plane substrate of Si of refractive index $n'' = 3.85 + i0.018$; the spheres are assumed to be embedded in vacuo ($n' = 1$). In the lower part of the figure we report the scattered intensity from a dispersion of the binary aggregates of the spheres described above with random distribution of their orientations. In

the left part of the figure are shown, in μm^2, the quantities $r^2 I_{\varphi\varphi}/I_0$ for the single spheres and $r^2\langle I_{\varphi\varphi}\rangle/I_0$ for the aggregates, where I_0 is the intensity of the incident field and the angular bracketts denote orientational average. In the right part of the figure the reported quantities are $r^2 I_{\varphi\vartheta}/I_0$ for the single spheres and $r^2\langle I_{\varphi\vartheta}\rangle/I_0$ for the aggregates as an example of cross-polarization.



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At first sigth the patterns of the co-polarized intensity from single spheres and from orientationally averaged aggregates look rather similar. Actually, the intensity at the main peak is larger for the case of the aggregates, as it was expected because an aggregate contains twice the refractive material of a single sphere. The main peak occurs in the direction of reflection both for single and for aggregated spheres ($\vartheta_S = 90°$, $\varphi_S = 135°$. Nevertheless the width of the peak from the aggregates is smaller that for the single spheres. This suggests that the effect of the multiple scattering processes that occur among the aggregated spheres are not suppressed by the averaging procedure. In any case a non-vanishing field propagates along the surface, although in the case of the aggregates, its intensity is lower that for single spheres. As for the cross-polarized intensity, we notice that the pattern from single spheres appears sharper than the one from aggregates. This effect that we remarked even in the case of a perfectly reflecting surface,[8] can be attributed to the averaging procedure.

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Differential light-scattering photometer using a CCD camera

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Optical scattering methods, based on the differential scattered light measurements, are extremely useful in particle sizing and characterization and, despite a large widespread of proposed experimental setups, there still exists a possibility to modify and improve them [1-4]. In this work we present an extension of a previously reported differential-light scattering photometer suitable for measurements over a wide angular range of scattered intensity coming from either aqueous solutions of spherical particles or particles deposited over a flat surface [5,6]. In this scheme, sketched in fig. 1, the light, coming from a linearly polarized 10 mW 632.8 nm He-Ne laser, is focused on the sample and the scattered portion is collected by an ellipsoidal mirror (Melles-Griot 02 REM 11), selected by a circularly shaped mask and by a diaphragm and, finally, brought directly to the CCD sensor of a common 8-bit black and white video camera. The scattered intensity is acquired as an 8-bit image through a PC acquisition card using DMA transfer over the PCI bus. The image has a circular ring shape and the angular intensity distribution is represented by the pixel intensity. In order to enlarge the measurement dynamic range, which would be intrinsically restricted to 8-bit, we used an I/O digital PC card to change automatically and in rapid succession the camera shutter opening times from 1/60 down to 1/10000 sec. In this way a nearly 15-bit dynamic range can be sampled.

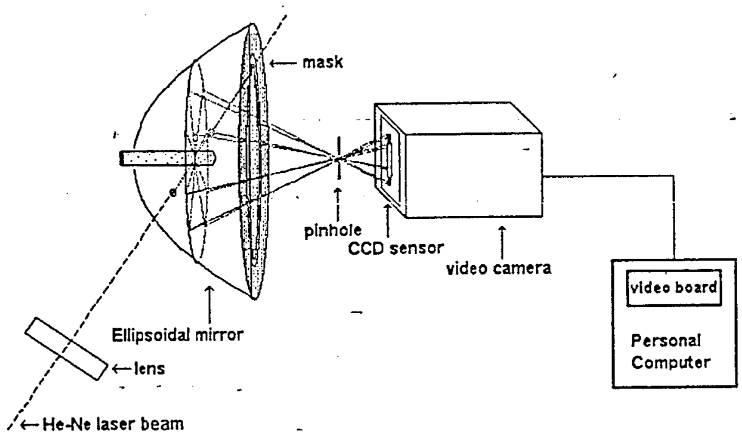

Fig. 1 - Schematic drawing of the experimental setup.

The present experimental setup offers the advantage, over the older one, of a much simpler configuration, having eliminated two optical components (i.e. two focalization lenses), and allows a wider scattering angle interval to be investigated. On the other hand its application is limited to static scattering experiments.

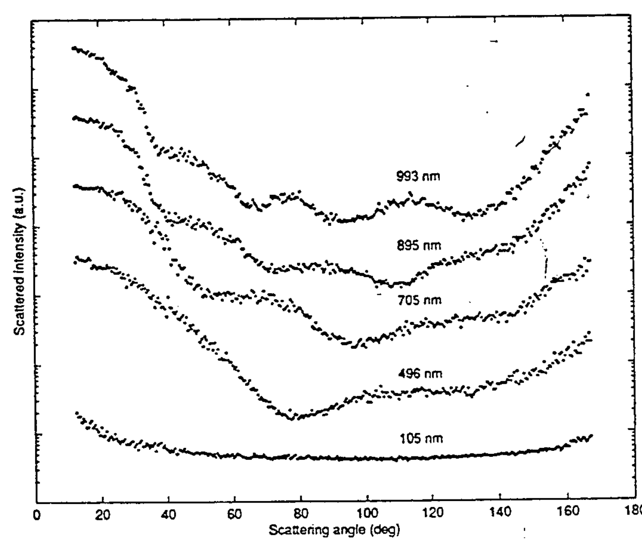

Fig. 2 - Measured scattered pattern for several particle diameters (curves are scaled for sake of clarity).

In order to test the apparatus effectiveness we carried out measurements on high purity, liquid-chromatography grade water solutions of polystyrene spheres ranging from 105

to 993 nm. The results, referred to a laser polarization perpendicular to the scattering plane, are reported in fig. 2. They show, for all sizes, an excellent agreement with both the previously measured ones and with the theoretical predictions which include geometrical corrections for both reflection and refraction processes [6,7].

This experimental setup is also suited for measurements of light scattered from particles on flat surfaces. In fact we deposited from a diluted solution 993 nm latex spheres on the specular surface of a conventional electronic grade silicon wafer. We checked the spheres distribution by optical microscopy, which excluded the presence of clusters and we estimated a number of nearly 40 spheres contained in the scattering area (a circle of 90 μm diameter). The preliminary results obtained for both perpendicular and parallel laser polarizations with respect to the scattering plane are reported in fig. 3.

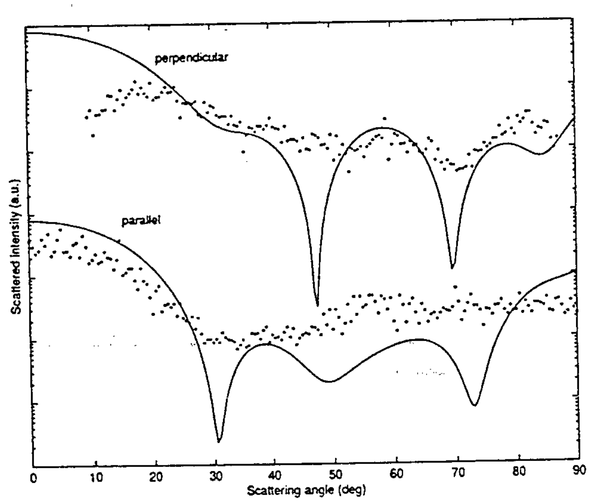

Fig. 3 - Measured (points) and theoretical scattered pattern for 993 nm sphere over a silicon surface for both perpendicular and parallel polarizations, with respect to the scattering plane, of the normal incident laser beam. Scattering angles are measured from surface normal.

For comparison we report also the theoretical patterns calculated on the basis of the electromagnetic field expansion into a series of spherical multipole fields [8]. As is clearly evident there is a poor agreement in the details, while the overall angular scattered intensity distribution seems to agree better. We believe this is mainly due to both the preliminary nature of the experimental measurements and to necessity to introduce some geometrical corrections to the pure theoretical scattered pattern.

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Optical properties of aggregated spheres in the vicinity of a plane surface

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Our previous theory for calculating the scattering pattern from single spheres in the vicinity of a dielectric substrate [J. Opt. Soc. Am. A 14,1505–1514 (1997)] is extended to the case of aggregated spheres. This extension preserves our main result that a non-vanishing field, though somewhat attenuated when the substrate is absorptive, can propagate along the interface; this feature is apparent in the patterns from all the aggregates that we considered. The effects that can be expected when scattering particles on the dielectric surface either aggregate or undergo some kind of subdivision are investigated by comparing the pattern from a sphere containing a chosen volume V of a given refractive material with the pattern from the aggregate of two such spheres and with the pattern from two aggregated spheres of the same material, each of volume $V/2$.

Accepted by JOSA A.

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1. Introduction

In the last years several approaches have been proposed for calculating the scattering pattern from surfaces seeded by particles of known morphology in view of the interest of this problem for pure research as well as for technological applications.[1–4] For the case of a metallic surface, that can be effectively modeled as a perfectly reflecting plane, image theory proved to be a suitable approach.[5] In practice, one has to solve, instead of the real problem, the equivalent problem of the scattering from the compound object that includes the actual particle and its image when they are illuminated by the superposition of the actual incident light and of the light that comes from the image source. The equivalent problem turns out to have computationally viable solutions for single and aggregated hemispheres with their flat face on the reflecting surface and for single and aggregated spheres,[6,7] as well as for cylinders with their axis parallel to the reflecting surface.[8]

Image theory is not easily extended to the case of a dielectric half-space[9] except when small particles are considered.[3] When the small-particle simplifications do not apply, the scattering problem can be alternatively solved by direct imposition of the appropriate boundary conditions both across the plane surface and across the surface of the particles. In this respect, the method devised by Bobbert and Vlieger[10] to deal with spheres deposited on a dielectric substrate is successful in solving exactly the boundary value problem without invoking any limitation of principle on the size of the particles to be dealt with. Nevertheless, these authors, in order to get the scattered field in the far zone, resort to an approximation that has rather severe effects on the reliability of their results: their procedure predicts, indeed, that no field propagates along the surface, contrary to the results of the experiments and of ab initio simulations.[11,12]

The difficulties of the approach of Bobbert and Vlieger have been overcome by the present authors through the formulation of a general rule for the reflection of a vector multipole field on a plane surface.[13] This rule, when applied to the description of the scattering from a sphere on or near a dielectric substrate,[14] yields a conceptually simple procedure to impose the boundary conditions also across the surface of the sphere and to get for the scattered field the correct behavior at infinity. The results that stem from this procedure are in agreement with the experiment and with the simulations[11,12] and predict, in particular, the propagation of a non-vanishing field along the surface.

The purpose of the present paper is to extend the procedure of Ref. 14 to the calculation of the scattering pattern from aggregated spheres on or near a dielectric surface. The need for such an extension is easily understood when one thinks that aggregation phenomena often occur among the particles on a substrate. In fact, aggregated spheres give origin to anisotropic particles whose scattering patterns are expected to be rather different from the patterns from single spheres and to depend on the orientation of the aggregate with respect to the incident field. In this respect we recall that the pattern of the scattered intensity from a low-density monolayer of identical particles all oriented alike but randomly distributed on the surface is merely proportional to the pattern from a single particle;[15] the case of a monolayer of particles whose orientations are randomly distributed around an axis orthogonal to the surface requires instead to consider the average over the orientational distribution. Since this averaging procedure implies further complications of formalism with respect to the case of fixed orientation, in this paper we deal with the latter case only and defer the problem of the orientational average to a further paper that is presently in progress.

In Section 2 we sketch the theory on which the calculation of the field scattered by an aggregate of spherical scatterers on the surface is based. Then, we discuss how the full pattern of the scattered intensity can be calculated for aggregates of fixed orientation. Further details on the mathematics of our procedure as well as on the related formalism and notation can be found in Refs. 13 and 14.

In Section 3 we present our specific results for binary aggregates, the scattering from whose component spheres was already dealt with by other authors with the help of suitable approximations.[16,17] We also present our results for the binary aggregates of spheres whose radius is chosen so that the total volume of the aggregate equals the volume of one of the spheres referred to above.

A few concluding remarks will be drawn in Section 4.

2. Theory

A. Statement of the problem

Let us assume that a plane surface separates a semi-infinite homogeneous isotropic medium of real refractive index n' from another semi-infinite isotropic homogeneous medium of (possibly complex) refractive index n'': hereafter we will refer to the region filled by the former medium as the accessible half-space. We also assume that a plane wave, of circular frequency ω, propagates within the accessible half-space and illuminates a cluster of spheres, i. e. a group of spherical scatterers whose mutual distances are so small that dependent scattering need to be considered. In order to calculate the scattering pattern from such an object we need to determine the total field at any point P in the accessible half-space: we will refer to the total field as $\mathbf{E}^{Int}$ when P lies within any of the spheres that compose the cluster, otherwise we will refer to the field as $\mathbf{E}^{Ext}$. Now, if no particle were present, the total field, $\mathbf{E}^{Ext}$, would be the superposition of the incident wave, $\mathbf{E}^{I}$, and of the field that has been reflected by the surface, $\mathbf{E}^{R}$; these fields are related to each other by the reflection conditions on the plane surface. When a cluster is present we need to add to $\mathbf{E}^{I}$ and $\mathbf{E}^{R}$ the field that is scattered by the cluster itself, $\mathbf{E}^{S}$, as well as the field that, after scattering by the cluster, is reflected by the plane surface, $\mathbf{E}^{RS}$. Ultimately

$$\mathbf{E}^{Ext} = \mathbf{E}^{I} + \mathbf{E}^{R} + \mathbf{E}^{S} + \mathbf{E}^{RS}, \tag{1}$$

and $\mathbf{E}^{S}$ is determined by imposing to $\mathbf{E}^{Ext}$ the appropriate boundary conditions across the surface of each of the spheres in the cluster. In this respect we recall that, on the assumption that all the fields depend on time through the factor $\exp(-i\omega t)$, the magnetic field $\mathbf{B}$ that is also needed to impose the boundary conditions is given by the Maxwell equation

$$i\mathbf{B} = \frac{1}{k}\nabla \times \mathbf{E},$$

with $k = \omega/c$, that holds true both for $\mathbf{B}^{Int}$ and $\mathbf{B}^{Ext}$.

The geometry that we adopt for our study is depicted in Fig. 1. The accessible half-space coincides with the region $z < 0$ of a cartesian frame of reference, of origin O, whose z axis is characterized by the unit vector $\hat{\mathbf{z}}$: the surface on which the reflection occurs is thus the plane $z = 0$. We assume that the center of the spheres, of radius ρ_α and refractive index n_α, that compose the aggregate lie at the points O'_α whose vector position will be denoted with $\mathbf{R}'_\alpha$. It is also convenient to define the points O''_α that are the images of the centers of the spheres with respect to the surface and to denote their vector positions as $\mathbf{R}''_\alpha$. The vector position of a point P will be denoted as $\mathbf{r}$ with respect to O, as $\mathbf{r}'_\alpha = \mathbf{r} - \mathbf{R}'_\alpha$ with respect to O'_α and as $\mathbf{r}''_\alpha = \mathbf{r} - \mathbf{R}''_\alpha$ with respect to O''_α.

B. Multipole expansion of the field

The electromagnetic plane wave

$$\mathbf{E}^{I}_{\eta} = E_{0\eta}\hat{\mathbf{u}}_{I\eta}\exp(i\mathbf{k}_I \cdot \mathbf{r}),$$

that propagates through the halfspace $z < 0$, is reflected into the plane wave

$$\mathbf{E}^{R}_{\eta} = E'_{0\eta}\hat{\mathbf{u}}_{R\eta}\exp(i\mathbf{k}_R \cdot \mathbf{r}),$$

where $\mathbf{k}_I = n'k\hat{\mathbf{k}}_I$ and $\mathbf{k}_R = n'k\hat{\mathbf{k}}_R$ are the propagation vectors of the incident and of the reflected plane wave and $\hat{\mathbf{u}}_{I\eta}$ and $\hat{\mathbf{u}}_{R\eta}$, are the respective unit polarization vectors whose index η distinguish

whether they are parallel ($\eta = 1$) or perpendicular ($\eta = 2$) to the plane of incidence, i. e. to the plane that contains $\mathbf{k}_I$, $\mathbf{k}_R$ and the z axis. Our actual choice for the polarization vectors is

$$\hat{\mathbf{u}}_{I1} \times \hat{\mathbf{u}}_{I2} = \hat{\mathbf{k}}_I, \quad \hat{\mathbf{u}}_{R1} \times \hat{\mathbf{u}}_{R2} = \hat{\mathbf{k}}_R$$

with $\hat{\mathbf{u}}_{R2} \equiv \hat{\mathbf{u}}_{I2}$. The reflection condition yields the relation

$$E'_{0\eta} = F_\eta(\vartheta_I) E_{0\eta},$$

where ϑ_I is the angle between $\mathbf{k}_I$ and $\hat{\mathbf{z}}$ and the quantities $F_\eta(\vartheta_I)$ are the Fresnel coefficients[18] for the reflection of a plane wave with polarization along $\hat{\mathbf{u}}_{I\eta}$

$$F_1(\vartheta_I) = \frac{n^2 \cos\vartheta_I - \beta}{n^2 \cos\vartheta_I + \beta}, \quad F_2(\vartheta_I) = \frac{\cos\vartheta_I - \beta}{\cos\vartheta_I + \beta},$$

where $n = n''/n'$ and

$$\beta = \left[(n^2 - 1) + \cos^2\vartheta_I\right]^{1/2}.$$

Accordingly, we rewrite the reflected plane wave as

$$\mathbf{E}^R_\eta = F_\eta(\vartheta_I) E_{0\eta} \hat{\mathbf{u}}_{R\eta} \exp(i\mathbf{k}_R \cdot \mathbf{r}).$$

For our purposes it is convenient to expand both $\mathbf{E}^I_\eta$ and $\mathbf{E}^R_\eta$ in terms of vector multipole fields. With reference to the origin O such expansions read

$$\mathbf{E}^I_\eta = E_{0\eta} \sum_{plm} \mathbf{J}^{(p)}_{lm}(\mathbf{r}, n'k) W^{(p)}_{lm}(\hat{\mathbf{u}}_{I\eta}, \hat{\mathbf{k}}_I), \tag{2}$$

$$\mathbf{E}^R_\eta = F_\eta(\vartheta_I) E_{0\eta} \sum_{plm} \mathbf{J}^{(p)}_{lm}(\mathbf{r}, n'k) W^{(p)}_{lm}(\hat{\mathbf{u}}_{R\eta}, \hat{\mathbf{k}}_R), \tag{3}$$

where we define the vector multipole fields[18]

$$\mathbf{J}^{(1)}_{lm}(\mathbf{r}, K) = j_l(Kr)\mathbf{X}_{lm}(\hat{\mathbf{r}}), \quad \mathbf{J}^{(2)}_{lm}(\mathbf{r}, K) = \frac{1}{K}\nabla \times j_l(Kr)\mathbf{X}_{lm}(\hat{\mathbf{r}}) \tag{4}$$

and the amplitudes[13]

$$W^{(p)}_{lm}(\hat{\mathbf{u}}, \hat{\mathbf{K}}) = 4\pi i^{p+l-1}(-)^{m+1}\mathbf{Z}^{(p)}_{l,-m}(\hat{\mathbf{K}}) \cdot \hat{\mathbf{u}}.$$

In the preceding equations the functions $\mathbf{X}_{lm}$ are vector spherical harmonics,[18]

$$\mathbf{Z}^{(1)}_{lm}(\hat{\mathbf{K}}) = \mathbf{X}_{lm}(\hat{\mathbf{K}}), \quad \mathbf{Z}^{(2)}_{lm}(\hat{\mathbf{K}}) = \mathbf{X}_{lm}(\hat{\mathbf{K}}) \times \hat{\mathbf{K}}. \tag{5}$$

are the transverse vector harmonics and the superscript $p = 1, 2$ is a parity index that distinguishes the magnetic multipoles ($p = 1$) from the electric ones ($p = 2$). The reflection condition yields the relation[7]

$$W^{(p)}_{\eta lm}(\hat{\mathbf{u}}_{R\eta}, \hat{\mathbf{k}}_R) = (-)^{\eta+p+l+m} W^{(p)}_{\eta lm}(\hat{\mathbf{u}}_{I\eta}, \hat{\mathbf{k}}_I),$$

so that the amplitudes of $\mathbf{E}^R_\eta$ never need to be calculated explicitly.

Since $\mathbf{E}^I_\eta$ and $\mathbf{E}^R_\eta$ must satisfy the boundary conditions also across the surface of each of the scattering spheres, we rewrite their expansions, Eqs. (2) and (3), in terms of spherical vector multipole fields with origin at the center of the α-th sphere, O'_α, as

$$\begin{aligned}\mathbf{E}_\eta^I &= E_{0\eta}\hat{\mathbf{u}}_{I\eta}\exp[i\mathbf{k}_I\cdot(\mathbf{R}'_\alpha+\mathbf{r}'_\alpha)]\\ &=\exp(i\mathbf{k}_I\cdot\mathbf{R}'_\alpha)E_{0\eta}\sum_{plm}\mathbf{J}_{lm}^{(p)}(\mathbf{r}'_\alpha,n'k)W_{lm}^{(p)}(\hat{\mathbf{u}}_{I\eta},\hat{\mathbf{k}}_I),\end{aligned}\tag{6}$$

$$\begin{aligned}\mathbf{E}_\eta^R &= F_\eta(\vartheta_I)E_{0\eta}\hat{\mathbf{u}}_{R\eta}\exp[i\mathbf{k}_R\cdot(\mathbf{R}'_\alpha+\mathbf{r}'_\alpha)]\\ &=\exp(i\mathbf{k}_R\cdot\mathbf{R}'_\alpha)F_\eta(\vartheta_I)E_{0\eta}\sum_{plm}\mathbf{J}_{lm}^{(p)}(\mathbf{r}'_\alpha,n'k)W_{lm}^{(p)}(\hat{\mathbf{u}}_{R\eta},\hat{\mathbf{k}}_R).\end{aligned}\tag{7}$$

The field that is scattered by an aggregate of spheres embedded within a homogeneous medium can always be described as the superposition of the fields that are scattered by each one of the spheres. Since the latter fields must satisfy the radiation condition at infinity we write

$$\mathbf{E}_\eta^S = E_{0\eta}\sum_\alpha\sum_{plm}\mathbf{H}_{lm}^{(p)}(\mathbf{r}'_\alpha,n'k)A_{\eta\alpha lm}^{(p)},\tag{8}$$

where the multipole fields $\mathbf{H}$ are identical to the $\mathbf{J}$ fields, Eq. (4), except for the substitution of the spherical Hankel functions of the first kind h_l for the Bessel functions j_l; the amplitudes A, that are as yet unknown, are determined by the boundary conditions at the surface of each of the component spheres.[19]

We still need to consider the field $\mathbf{E}_\eta^{RS}$ that, after scattering by the cluster, is reflected by the surface. According to our previous results,[13,14] at any point of the accessible half-space, $\mathbf{E}_\eta^{RS}$ can be written as

$$\mathbf{E}_\eta^{RS} = E_{0\eta}\sum_\alpha\sum_{plm}\mathbf{H}_{lm}^{(p)}(\mathbf{r}''_\alpha,n'k)\overline{A}_{\eta\alpha lm}^{(p)},\tag{9}$$

i. e. as a superposition of $\mathbf{H}$ multipole fields with origin at the image points O''_α. The amplitudes $\overline{A}_{\eta\alpha lm}^{(p)}$ are related to the amplitudes of the scattered field $\mathbf{E}^S$ by the equation

$$\overline{A}_{\eta\alpha lm}^{(p)} = \sum_{p'l'}a_{\alpha;l,l';m}^{(p,p')}A_{\eta\alpha l'm}^{(p')}.\tag{10}$$

The explicit expression of the amplitudes $a_{\alpha;l,l';m}^{(p,p')}$ is

$$a_{\alpha;l,l';m}^{(p,p')} = \sum_{p''l''}(\mathcal{H}_\alpha^{-1})_{l,l'';m}^{(p,p'')}\mathcal{F}_{\alpha;l'',l';m}^{(p'',p')},$$

where $\mathcal{H}_\alpha^{-1}$ is the inverse to the matrix $\mathcal{H}(\overline{\mathbf{R}}_{\alpha\alpha},n'k)$ that effects the transfer of the origin of the $\mathbf{H}$ multipole fields from O''_α to O'_α and $\overline{\mathbf{R}}_{\alpha\alpha}=\mathbf{R}'_\alpha-\mathbf{R}''_\alpha$. In turn, the quantities $\mathcal{F}_{\alpha;l'',l';m}^{(p'',p')}$, are the elements of the matrix $\mathcal{F}_\alpha$ that effects the reflection on the plane surface of the $\mathbf{H}$ multipole fields with origin at O'_α.[13,14]

The last expansion that we need to consider it that of the field within the spheres of the cluster. Since the field within the α-th sphere must be regular at O'_α its expansion can be taken in the form

$$\mathbf{E}_\eta^{Int} = E_{0\eta}\sum_{plm}\mathbf{J}_{lm}^{(p)}(\mathbf{r}'_\alpha,n_\alpha k)C_{\eta\alpha lm}^{(p)}.\tag{11}$$

C. Amplitudes of the scattered field

In order to calculate the amplitudes $A_{\eta\alpha lm}^{(p)}$ by imposing the boundary conditions across the surface of each of the spheres that compose the cluster, a few steps are still necessary. When we come

to consider the α-th sphere, the fields $\mathbf{E}_\eta^I$, Eq. (6), and $\mathbf{E}_\eta^R$, Eq. (7), that contribute to $\mathbf{E}_\eta^{Ext}$, Eq. (1), as well as $\mathbf{E}_\eta^{Int}$, Eq. (11), are all given in terms of vector multipole fields with origin at O'_α. The scattered field $\mathbf{E}_\eta^S$, Eq. (8), and the reflected scattered field $\mathbf{E}_\eta^{RS}$, Eq. (9), that are given by superpositions of vector multipole fields with different origins, can also be expressed in terms of multipole fields with origin at O'_α through the use of the appropriate addition theorem.[20] The result for the scattered field $\mathbf{E}_\eta^S$ is[13]

$$\mathbf{E}_\eta^S = E_{0\eta} \sum_{plm} \Big[\mathbf{H}_{lm}^{(p)}(\mathbf{r}'_\alpha, n'k) A_{\eta\alpha lm}^{(p)} + \mathbf{J}_{lm}^{(p)}(\mathbf{r}'_\alpha, n'k) \sum_{p'l'm'} \sum_{\beta\neq\alpha} \mathcal{H}_{lm,l'm'}^{(p,p')}(\mathbf{R}'_{\beta\alpha}, n'k) A_{\eta\beta l'm'}^{(p')} \Big],$$

where $\mathbf{R}'_{\beta\alpha} = \mathbf{R}'_\alpha - \mathbf{R}'_\beta$, while for the reflected scattered field $\mathbf{E}^{RS}$ we obtain

$$\mathbf{E}_\eta^{RS} = E_{0\eta} \sum_{plm} \mathbf{J}_{lm}^{(p)}(\mathbf{r}'_\alpha, n'k) \sum_{p'l'm'} \Big[\mathcal{H}_{lm,l'm'}^{(p,p')}(\overline{\mathbf{R}}_{\alpha\alpha}, n'k) \overline{A}_{\eta\alpha l'm'}^{(p')} + \sum_{\beta\neq\alpha} \mathcal{H}_{lm,l'm'}^{(p,p')}(\overline{\mathbf{R}}_{\beta\alpha}, n'k) \overline{A}_{\eta\beta l'm'}^{(p')} \Big],$$

where $\overline{\mathbf{R}}_{\beta\alpha} = \mathbf{R}'_\alpha - \mathbf{R}''_\beta$. Using the expression, Eq. (10), for the amplitudes $\overline{A}$ and taking into account that $\mathcal{H}_{lm,l'm'}^{(p,p')}(\overline{\mathbf{R}}_{\alpha\alpha}, n'k) = \mathcal{H}_{l,l';m}^{(p,p')}(\overline{\mathbf{R}}_{\alpha\alpha}, n'k)\delta_{mm'}$ we get the final result

$$\mathbf{E}_\eta^{RS} = E_{0\eta} \sum_{plm} \mathbf{J}_{lm}^{(p)}(\mathbf{r}'_\alpha, n'k) \sum_{p'l'} \Big[\mathcal{F}_{\alpha;l,l';m}^{(p,p')} A_{\eta\alpha l'm}^{(p')} + \sum_{\beta\neq\alpha} \sum_{p''l''m'} \mathcal{Q}_{lm,l''m'}^{(p,p'')}(\overline{\mathbf{R}}_{\beta\alpha}, n'k) \mathcal{F}_{\beta;l'',l';m'}^{(p'',p')} A_{\eta\beta l'm'}^{(p')} \Big],$$

where we define

$$\mathcal{Q}_{lm,l''m'}^{(p,p'')}(\overline{\mathbf{R}}_{\beta\alpha}, n'k) = \sum_{p'l'} \mathcal{H}_{lm,l'm'}^{(p,p')}(\overline{\mathbf{R}}_{\beta\alpha}, n'k) \big(\mathcal{H}_\beta^{-1}\big)_{l',l'';m'}^{(p',p'')}.$$

We are now ready to impose the boundary conditions across the surface of each of the scattering spheres by applying to $\mathbf{E}$ and $\mathbf{B}$ the procedure that we described elsewhere in full detail.[19] This procedure yields, for each α, p, l and m, four equations among which the amplitudes of the internal field C can be easily eliminated. As a result we get for the amplitudes $A_{\eta\alpha lm}^{(p)}$ the system of linear non-homogeneous equations

$$\sum_\beta \sum_{p'l'm'} \big(\mathcal{M}^{-1}\big)_{\alpha lm,\beta l'm'}^{(p,p')} A_{\eta\beta l'm'}^{(p')} = -\mathcal{W}_{\eta\alpha lm}^{(p)}, \tag{12}$$

where

$$\begin{aligned}\big(\mathcal{M}^{-1}\big)_{\alpha lm,\beta l'm'}^{(p,p')} = {} & \big(R^{-1}\big)_{\alpha l}^{(p)} \delta_{\alpha\beta}\delta_{pp'}\delta_{ll'}\delta_{mm'} \\ & + \mathcal{H}_{lm,l'm'}^{(p,p')}(\mathbf{R}'_{\beta\alpha}, n'k) + \mathcal{F}_{\alpha;l,l';m}^{(p,p')}\delta_{\alpha\beta}\delta_{mm'} \\ & + \sum_{p''l''} \mathcal{Q}_{lm,l''m'}^{(p,p'')}(\overline{\mathbf{R}}_{\beta\alpha}, n'k) \mathcal{F}_{\beta;l'',l';m'}^{(p'',p')},\end{aligned}$$

with

$$R_{\alpha l}^{(p)} = \frac{(1+\overline{n}_\alpha\delta_{p1})u_l'(n_\alpha k\rho_\alpha)u_l(n'k\rho_\alpha) - (1+\overline{n}_\alpha\delta_{p2})u_l(n_\alpha k\rho_\alpha)u_l'(n'k\rho_\alpha)}{(1+\overline{n}_\alpha\delta_{p1})u_l'(n_\alpha k\rho_\alpha)w_l(n'k\rho_\alpha) - (1+\overline{n}_\alpha\delta_{p2})u_l(n_\alpha k\rho_\alpha)w_l'(n'k\rho_\alpha)}$$

and

$$\overline{n}_\alpha = \frac{n_\alpha}{n'} - 1, \quad u_l(x) = xj_l(x), \quad w_l(x) = xh_l(x).$$

The quantities $R_{\alpha l}^{(1)}$ and $R_{\alpha l}^{(2)}$ coincide with the Mie coefficients b_l and a_l, respectively,[21] for a homogeneous sphere of refractive index n_α embedded into a homogeneous medium of refractive index n'. In turn

$$\mathcal{W}_{\eta\alpha lm}^{(p)} = \exp(i\mathbf{k}_I \cdot \mathbf{R}_\alpha')W_{lm}^{(p)}(\hat{\mathbf{u}}_{I\eta}, \hat{\mathbf{k}}_I) + F_\eta(\vartheta_I)\exp(i\mathbf{k}_R \cdot \mathbf{R}_\alpha')W_{lm}^{(p)}(\hat{\mathbf{u}}_{R\eta}, \hat{\mathbf{k}}_R).$$

D. Scattered intensity

Once the amplitudes $A_{\eta\alpha lm}^{(p)}$ of $\mathbf{E}_\eta^S$ have been calculated by solving Eq. (12), the reflected-scattered field, $\mathbf{E}_\eta^{RS}$, is also determined by Eqs. (9) and (10). Therefore, the superposition of $\mathbf{E}_\eta^S$, Eq. (8), and of $\mathbf{E}_\eta^{RS}$, Eq. (9), yields the field that would be observed by an optical instrument at any point in the accessible half-space. Hereafter we disregard the reflected field $\mathbf{E}_\eta^R$ because it can be observed in the direction of reflection only. Furthermore, it is convenient to use again the addition theorem of ref. 20 to refer both $\mathbf{E}_\eta^S$ and $\mathbf{E}_\eta^{RS}$ to a common origin that we choose to be the point O that is defined in Fig. 1. Accordingly, the field that would be observed in the far zone of the accessible half-space takes on the form

$$\mathbf{E}_\eta^{Obs} = E_{0\eta}\sum_{plm}\mathbf{H}_{lm}^{(p)}(\mathbf{r}, n'k)\mathcal{A}_{\eta lm}^{(p)},$$

with

$$\mathcal{A}_{\eta lm}^{(p)} = \sum_\alpha\sum_{p'l'm'}\left[\mathcal{J}_{lm,l'm'}^{(p,p')}(-\mathbf{R}_\alpha', n'k)A_{\eta\alpha l'm'}^{(p')} + \mathcal{J}_{lm,l'm'}^{(p,p')}(-\mathbf{R}_\alpha'', n'k)\overline{A}_{\eta\alpha l'm'}^{(p')}\right], \tag{13}$$

where the quantities $\mathcal{J}$ are the elements of the matrix that effects the transfer of origin of the $\mathbf{J}$ multipole fields.[13] Then, on account of the asymptotic form of the $\mathbf{H}$ fields for large values of $n'kr$ and of their transversality in the far zone, we are led to write $\mathbf{E}_\eta^{Obs}$ as

$$\mathbf{E}_\eta^{Obs} = \frac{\exp(in'kr)}{r}E_{0\eta}\mathbf{f}_\eta,$$

where we define the normalized scattering amplitude[18] $\mathbf{f}_\eta$ that, in terms of the transverse vector harmonics $\mathbf{Z}_{lm}^{(p)}(\hat{\mathbf{r}})$, Eq. (5), reads[14]

$$\mathbf{f}_\eta = \frac{1}{n'k}\sum_{plm}(-i)^{p+l}\mathbf{Z}_{lm}^{(p)}(\hat{\mathbf{r}})\mathcal{A}_{\eta lm}^{(p)}.$$

Therefore, the intensity that would be detected in the direction $\hat{\mathbf{k}}_O$ with polarization along $\hat{\mathbf{u}}_{O\eta'}$ is

$$I_{\eta'\eta} = \frac{1}{r^2}|E_{0\eta}f_{\eta'\eta}|^2 = \frac{1}{r^2}I_{0\eta}|f_{\eta'\eta}|^2,$$

where $I_{0\eta} = |E_{0\eta}|^2$ and

$$f_{\eta'\eta} = \mathbf{f}_\eta \cdot \hat{\mathbf{u}}_{O\eta'} = -\frac{i}{4\pi n' k} \sum_{plm} W_{lm}^{(p)*}(\hat{\mathbf{u}}_{O\eta'}, \hat{\mathbf{k}}_O) \mathcal{A}_{\eta lm}^{(p)}. \tag{14}$$

The efficiency of Eq. (14) can be improved when, as we assumed throughout, the refractive index n' is real. In this case, indeed, a consequence of the addition theorem of ref. 20 is the identity[13]

$$\exp(-i\mathbf{k} \cdot \mathbf{R}) W_{lm}^{(p)*}(\hat{\mathbf{u}}, \hat{\mathbf{k}}) = \sum_{p'l'm'} W_{l'm'}^{(p')*}(\hat{\mathbf{u}}, \hat{\mathbf{k}}) \mathcal{J}_{l'm',lm}^{(p',p)}(-\mathbf{R}, n'k), \tag{15}$$

where $\mathbf{k} = n'k\hat{\mathbf{k}}$. Now, with the help of Eq. (15), the multipole transfer elements $\mathcal{J}$ that are included into the definition of the amplitudes $\mathcal{A}$, Eq. (13), can be eliminated, so that Eq. (14) can be rewritten as

$$f_{\eta'\eta} = -\frac{i}{4\pi n' k} \sum_{\alpha} \sum_{plm} W_{lm}^{(p)*}(\hat{\mathbf{u}}_{O\eta'}, \hat{\mathbf{k}}_O)$$
$$\left[\exp(-i\mathbf{k}_O \cdot \mathbf{R}'_\alpha) A_{\eta\alpha lm}^{(p)} + \exp(-i\mathbf{k}_O \cdot \mathbf{R}''_\alpha) \overline{A}_{\eta\alpha lm}^{(p)}\right]. \tag{16}$$

Equation (16) is the one that we actually used in our calculations.

3. Results and discussion

The theory of Section 2 has been applied to the calculation of the full pattern of the scattered intensity from a binary aggregate of identical contacting spheres embedded in vacuo ($n' = 1$) and deposited on a substrate of Si ($n'' = 3.85 + i\,0.018$). We considered spheres of Si_3N_4 with refractive index $n_1 = 2.0$ and radius $\rho_1 = 450.0$ nm, illuminated by radiation of wavelength $\lambda = 632.8$ nm. The scattered intensity from single spheres of such a morphology deposited on the same substrate has already been calculated by Taubenblatt and Tran through the coupled-dipole method[16] and by Johnson[17] with the help of the Yousif-Videen approximation.[4,22] Our aim is thus to use our exact procedure to investigate the effect of the aggregation on scatterers whose properties are already well known. We also dealt with the pattern from binary aggregates of contacting spheres of Si_3N_4 with radius $\rho_2 = 357.0$ nm deposited on the surface because the total volume of such an aggregate equals that of a single sphere of radius ρ_1. This choice allows us to investigate also the effect of the subdivision of the material of a sphere. Hereafter we will refer to the aggregate of spheres of radius ρ_1 as the case of aggregation and to the aggregate of spheres of radius ρ_2 as the case of subdivision.

The geometry that we adopted to display our results is the one depicted in Fig. 1 of Ref. 6. Thus, the surface of the substrate coincides with the zx plane and the normal to the surface coincides with the y axis. When the single sphere is considered its center lies on the y axis while when the aggregate is considered the contacting point of the two spheres lies on the y axis and the axis of the aggregate is taken to be parallel either to the z axis or to the x axis. Nevertheless, the patterns that we actually report in this paper refer to the former case only because the patterns for the latter case do not present specific features that require a separate comment. The plane of incidence coincides with the xy plane and the direction of incidence forms an angle of 45° with the normal to the surface; more precisely, the polar angles of the direction of incidence are $\vartheta_I = 90°$ and $\varphi_I = 225°$. The incident light is polarized either along the meridians (ϑ-polarization) or along the parallels (φ-polarization) and both the φ-polarized and the ϑ-polarized component of the scattered wave are considered; in practice the quantity that we calculated is $\mathcal{I}_{\eta'\eta} = r^2 I_{\eta'\eta}/I_{0\eta}$ and both co-polarized ($\mathcal{I}_{\varphi\varphi}$ and $\mathcal{I}_{\vartheta\vartheta}$) and cross-polarized ($\mathcal{I}_{\vartheta\varphi}$ and $\mathcal{I}_{\varphi\vartheta}$) patterns are reported. The numerical results from which all the patterns were drawn achieve a convergency to four significant digits; this required to extend the multipole expansions in Sections 2 and 3 up to $l = 12$. We also recall that all the patterns that we are going to discuss have a number of features that do not depend on the system under study but rather stem from the boundary conditions and from the geometry of the

problem. The occurrence of these features that were discussed in Refs. 6 and 14 should be borne in mind for an appropriate interpretation of our results. Here we only recall that, since we consider the scattered field also out of the plane of incidence, cross-polarization effects are to be expected.[6]

In Fig. 2 we report our results for the pattern from a single sphere of radius ρ_1 in contact with the substrate. The section of the $\mathcal{I}_{\varphi\varphi}$ and of the $\mathcal{I}_{\vartheta\vartheta}$ patterns along the plane $\vartheta_I = 90°$ can be directly compared with the results of Taubenblatt and Tran[16] and of Johnson.[17] The differences that emerge from this comparison are easily explained along the lines of Ref. 14, where we stressed how the approximations used by other authors, possibly in conjunction with the approximation used by Bobbert and Vlieger[10] to get the far zone field, severely affect the reliability of the calculations. The results in Fig. 3 refer to a binary aggregate of spheres of radius ρ_1; the patterns from the aggregate of two spheres of radius ρ_2 are presented in Fig. 4.

We first notice that in agreement with our previous findings, the $\mathcal{I}_{\varphi\varphi}$ patterns show a non-vanishing field that propagates along the surface. Of course, its intensity is smaller than the intensity that we reported in our previous paper[14] but this is due to the fact that the refractive index of the substrate has a non-vanishing imaginary part.

To confirm this interpretation we compare the pattern of $\mathcal{I}_{\varphi\varphi}$ from a single sphere in Fig. 2a with the pattern from the same sphere on a perfectly reflecting surface. According to our calculations the latter pattern (not reported here) has a rounded shape in contrast to the rather peaked shape of the pattern in Fig. 2a. We also remark that also the patterns that we reported in Fig. 5 of Ref. 14 for spheres of different size and refractive index and for several choices of the (real) refractive index of the substrate present a rounded shape. We are thus led to conclude that a non-vanishing imaginary part of n'' produces a strong damping of the scattering patterns. As a result we are left with a noticeable peak around the direction of reflection ($\vartheta_S = 90°$, $\varphi_S = 135°$). This effect is not restricted to the case of a single sphere because the same peaked shape of the $\mathcal{I}_{\varphi\varphi}$ and of $\mathcal{I}_{\vartheta\vartheta}$ patterns can be observed in the case of the aggregated spheres; in other words this feature does not depend on the shape of the scatterers and should thus be due to the non-vanishing imaginary part of n''. The height of the main peaks, for φ-polarized incident field, depends more or less on the quantity of refractive material: for instance, the height of the $\mathcal{I}_{\varphi\varphi}$ peak is almost equal for the single sphere and for the case of subdivision whereas the height for the case of aggregation is about four times larger. Furthermore, the $\mathcal{I}_{\varphi\varphi}$ pattern for the case of aggregation show a noticeable increase, with respect to the case of subdivision, in the direction $\vartheta_S = 90°$, $\varphi_S = 180°$.

Anyway, the largest value of the scattered intensity does not occur exactly in the direction of reflection ($\vartheta_S = 90°$, $\varphi_S = 135°$). This may be surprising because, according to our calculations for the same particles in free space, the largest scattered intensity occurs in the forward direction that, for particles on a surface, coincides with the direction of reflection.[7] Actually, for the single sphere the maximum both of $\mathcal{I}_{\varphi\varphi}$ and $\mathcal{I}_{\vartheta\vartheta}$ occurs at $\varphi_S = 155°$, so that the coupling with the surface is so large as to produce a significant shift of the main peak with respect to the direction of reflection. For the aggregate of spheres of radius ρ_1 the maximum of $\mathcal{I}_{\varphi\varphi}$ occurs at $\varphi_S = 130°$ but it occurs at $\varphi_S = 150°$ for $\mathcal{I}_{\vartheta\vartheta}$; finally for the aggregate of spheres of radius ρ_2 the maximum of $\mathcal{I}_{\varphi\varphi}$ occurs again at $\varphi_S = 135°$ but the maximum of $\mathcal{I}_{\vartheta\vartheta}$ occurs at $\varphi_S = 150°$. Even though we do not report the specific patterns we can state that a similar behavior occurs even when the axis of the aggregates is parallel to the x axis. This seemingly erratic location of the maximum arises from the coupling of the scattering particles with the substrate and, for the case of aggregated spheres, also on the competing coupling of the component spheres among themselves. The strength of both these couplings is likely to differ for different polarization whereas the coupling among the component spheres is expected to depend on their size. Hence, the dependence of the location of the maximum on the polarization as well as on the radius of the aggregated spheres can be explained in terms of the ratio between these couplings.

Finally we notice that the $\mathcal{I}_{\varphi\varphi}$ and $\mathcal{I}_{\vartheta\vartheta}$ patterns from single spheres as well as from aggregated spheres both of radius ρ_1 and ρ_2 are of rather similar shape, even though the specific values of the scattered intensity may be noticeably different as we noted above with regard to the maximum. This means that an analysis of the scattered intensity in the plane of incidence only cannot yield enough information to discriminate the shape and size of the particles. In turn, even a cursory examination of the cross-polarized patterns $\mathcal{I}_{\vartheta\varphi}$ and $\mathcal{I}_{\varphi\vartheta}$ show that they depend both on the shape

and on the size of the scattering particles. We are thus led to conclude that any attempt to gain information on the morphology of particles deposited on a dielectric substrate requires considering both the co-polarized and the cross-polarized aspects of their scattering pattern.

4. Conclusions

The patterns that we discussed in Section 3 are only a sample of the changes undergone by the scattered intensity when spheres deposited on a dielectric substrate aggregate. However, the results that we reported prove that the present extension of the theory of Ref. 14 to aggregated spheres preserves the distinctive feature of the original approach that a non-vanishing field can propagate along the interface. We stress again that this feature is due to the fact that our procedure is based on no approximation, neither to get the far zone expression of the scattered field. Strictly speaking, our approach requires the truncation of the multipole expansions on which the formalism is based. Nevertheless, this unavoidable truncation is of numerical and not of physical nature. Therefore, once the convergency has been checked, our results remain physically reliable.

A noticeable advantage of any approach based on the multipole expansion of the field is the possibility of using the transformation properties of the vector multipole fields under rotation.[23] Once the scattering amplitude of an anisotropic scatterer has been calculated for a given orientation, producing the pattern for any other orientation is a fast and low-cost operation that produces a large amount of data.[6] Nevertheless, in the present paper we were interested in the effects that are due to the anisotropy of the aggregates so that we resolved to present the patterns for a specific choice of the orientation and of the direction of incidence, and to defer to a forthcoming paper the investigation of the patterns from randomly oriented aggregates.

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Fig. 1. Sketch of the geometry that we adopted in our theory. Only the α-th sphere of the cluster is shown for the sake of clarity.

Fig. 2. Pattern of the scattered intensity from the single sphere of radius ρ_1. The quantity that is actually reported (in μm^2) is $\mathcal{I}_{\varphi\varphi}$ in (a), $\mathcal{I}_{\vartheta\varphi}$ in (b), $\mathcal{I}_{\varphi\vartheta}$ in (c) and $\mathcal{I}_{\vartheta\vartheta}$ in (d).

Fig. 3. Pattern of the scattered intensity from the binary aggregate of the spheres of radius ρ_1. The quantity that is actually reported (in μm^2) is $\mathcal{I}_{\varphi\varphi}$ in (a), $\mathcal{I}_{\vartheta\varphi}$ in (b), $\mathcal{I}_{\varphi\vartheta}$ in (c) and $\mathcal{I}_{\vartheta\vartheta}$ in (d).

Fig. 4. Pattern of the scattered intensity from the binary aggregate of the spheres of radius ρ_2. The quantity that is actually reported (in μm^2) is $\mathcal{I}_{\varphi\varphi}$ in (a), $\mathcal{I}_{\vartheta\varphi}$ in (b), $\mathcal{I}_{\varphi\vartheta}$ in (c) and $\mathcal{I}_{\vartheta\vartheta}$ in (d).

Fig. 1

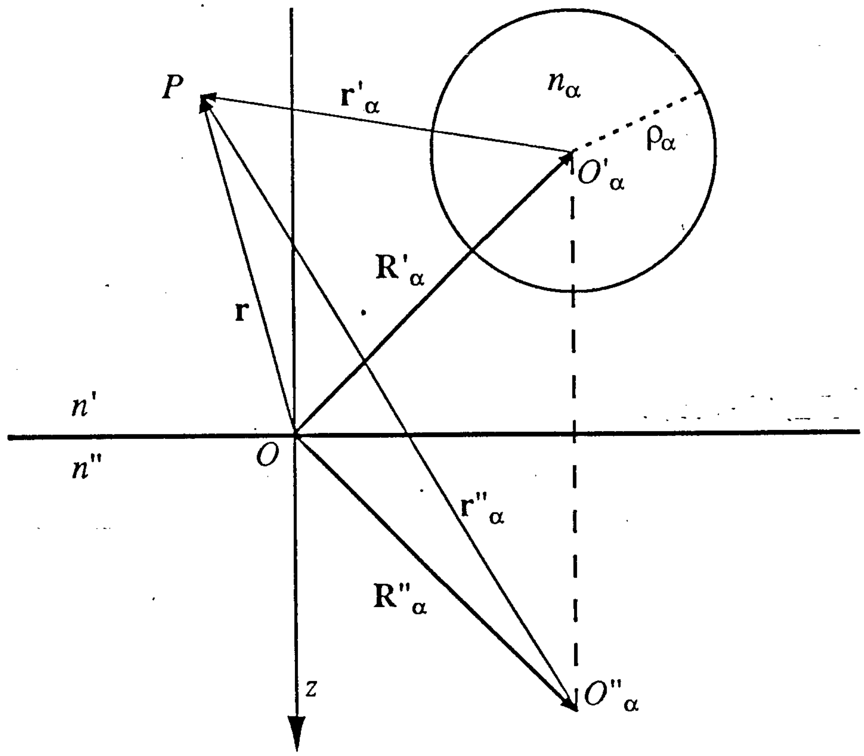

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Fig. 2a

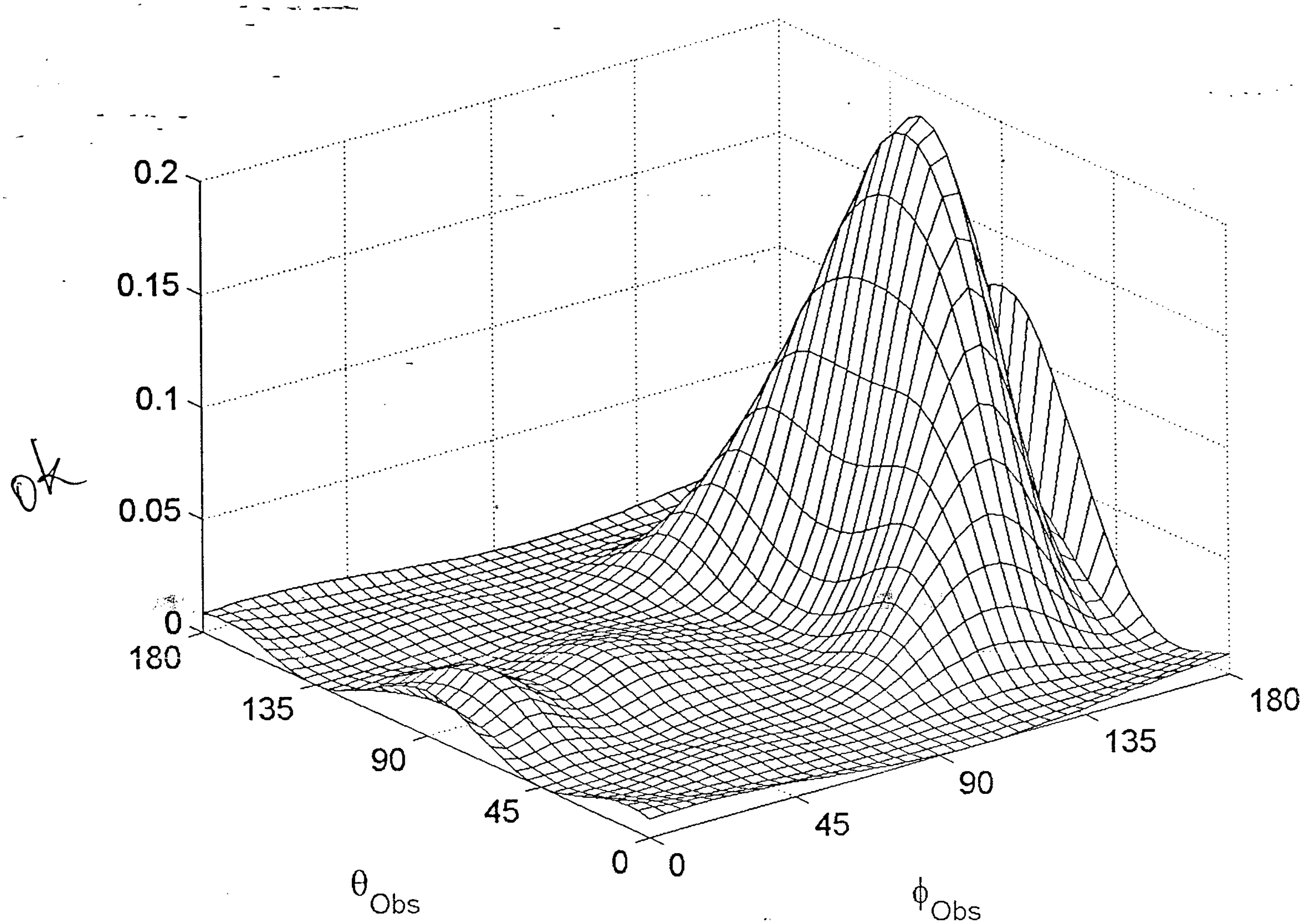
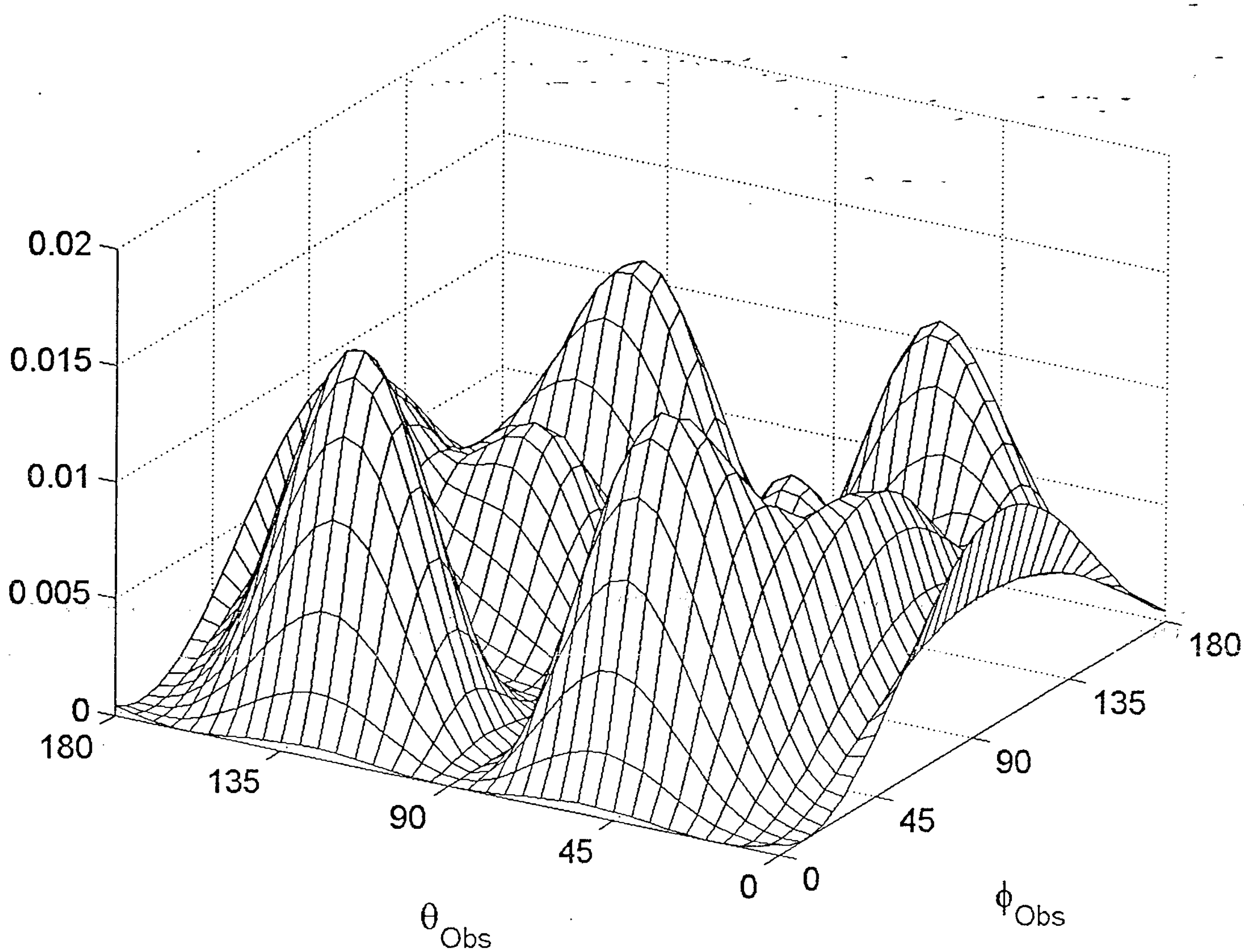


Fig 2b



0.02
0.015
0.01
0.005
0
180
135
90
45
0
θ_{Obs}
0
45
90
135
180
ϕ_{Obs}

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Fig. 2c

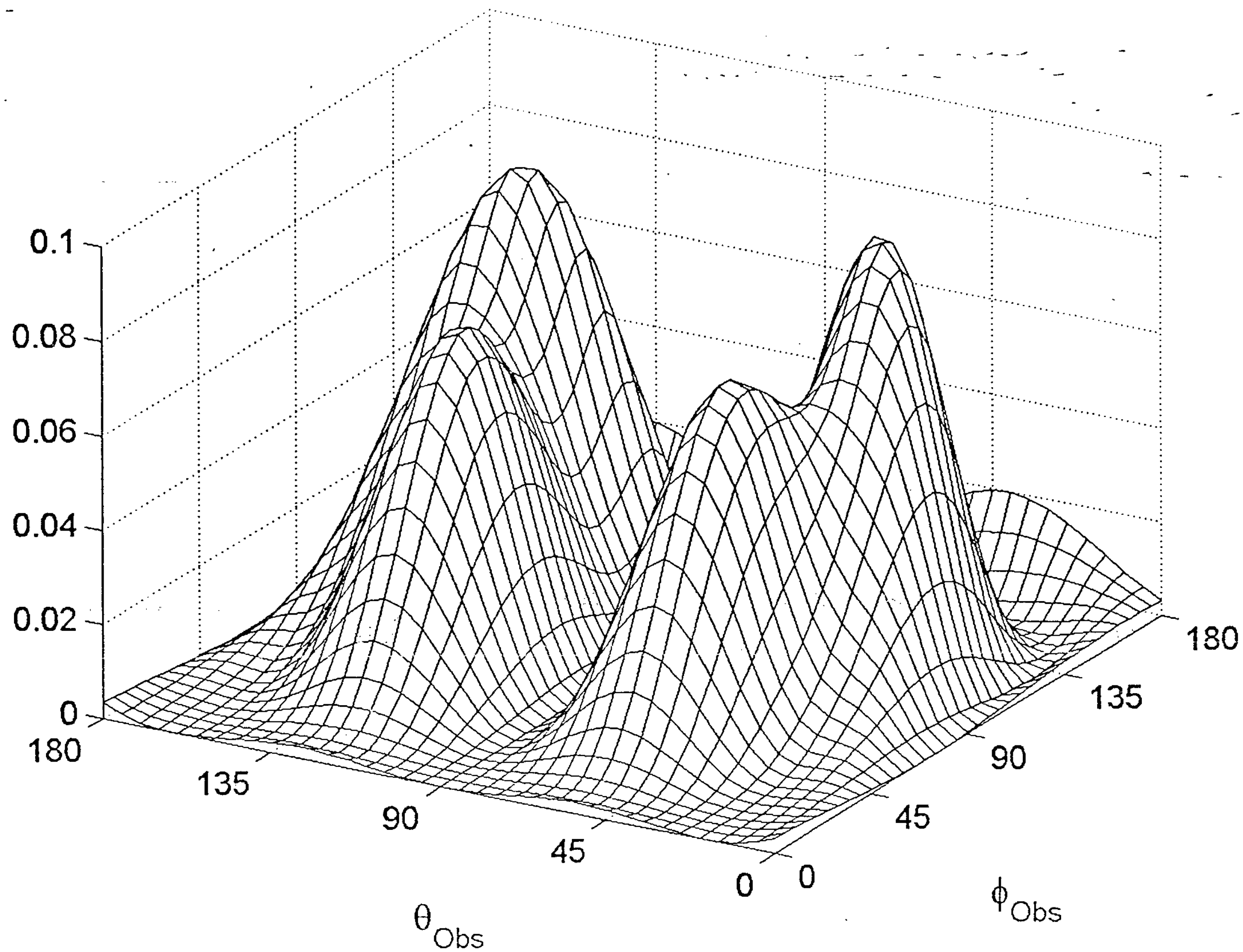


Fig 2d

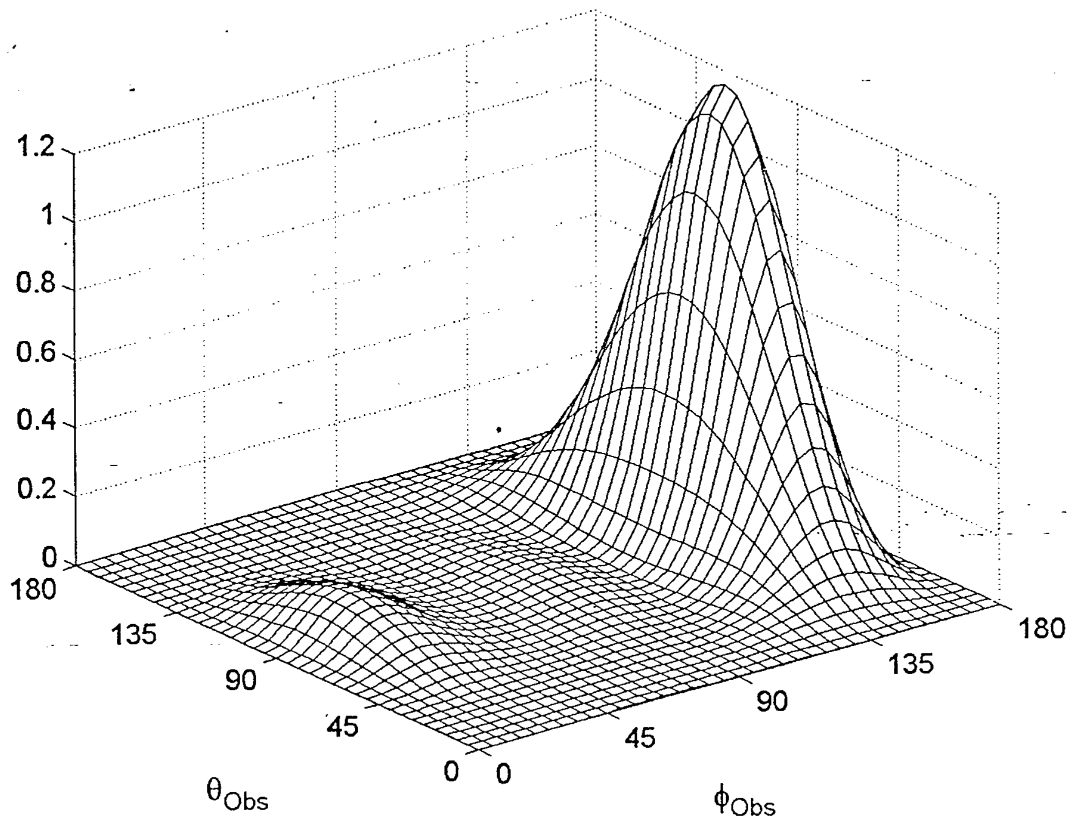
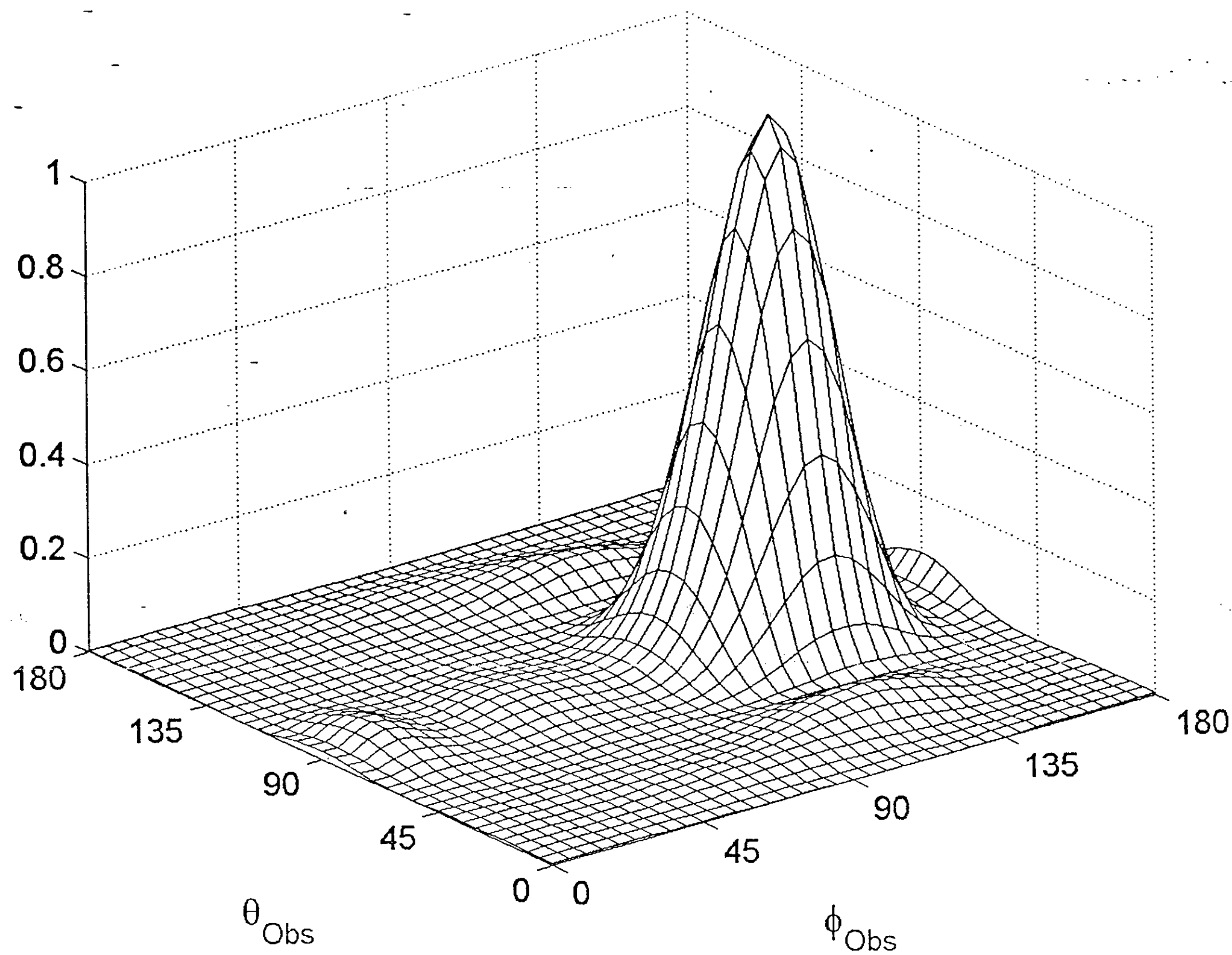


Fig. 3a



ok

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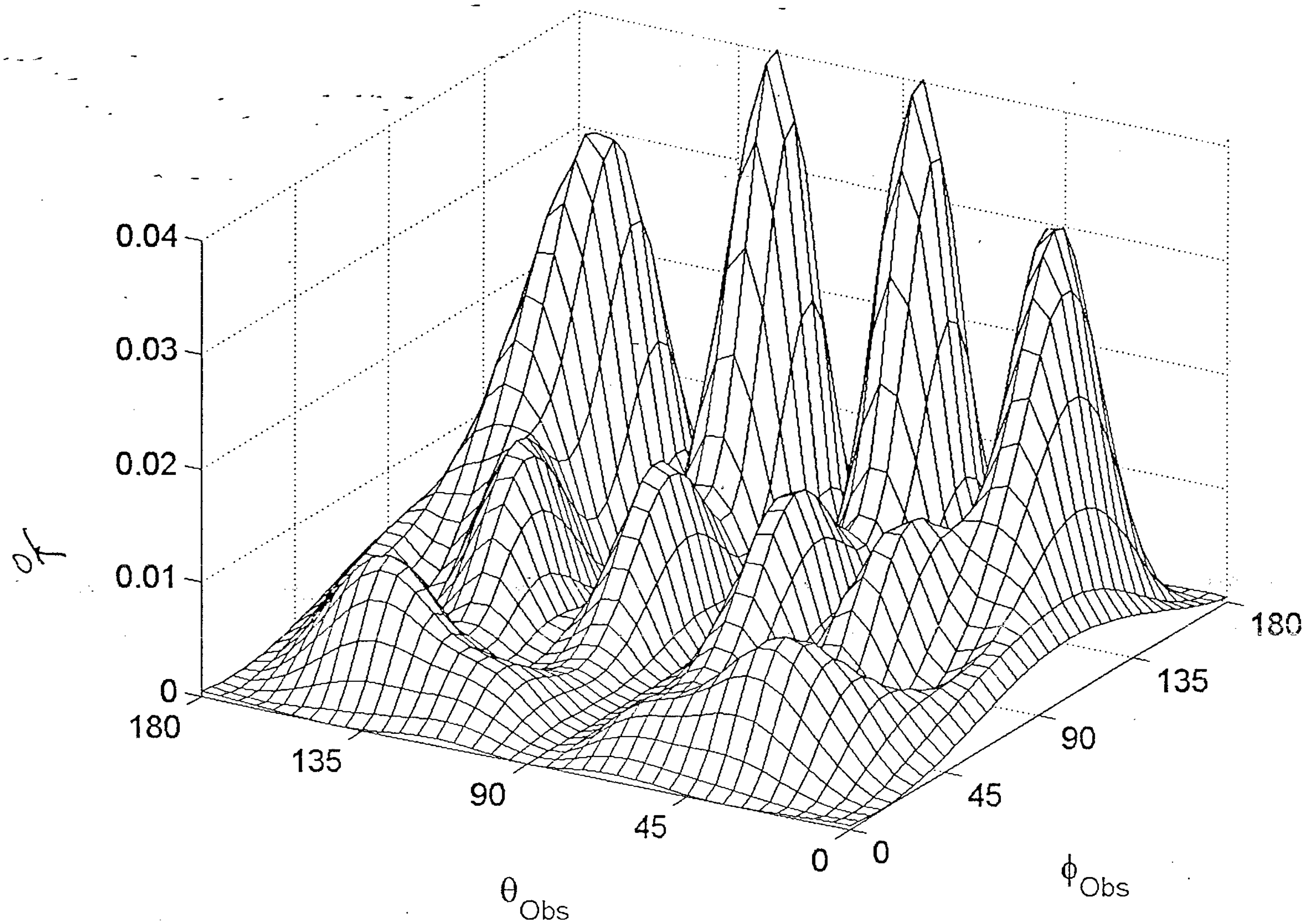
Fig. 3b



P. Deng et. al.: Optical Properties..

Fig. 3c

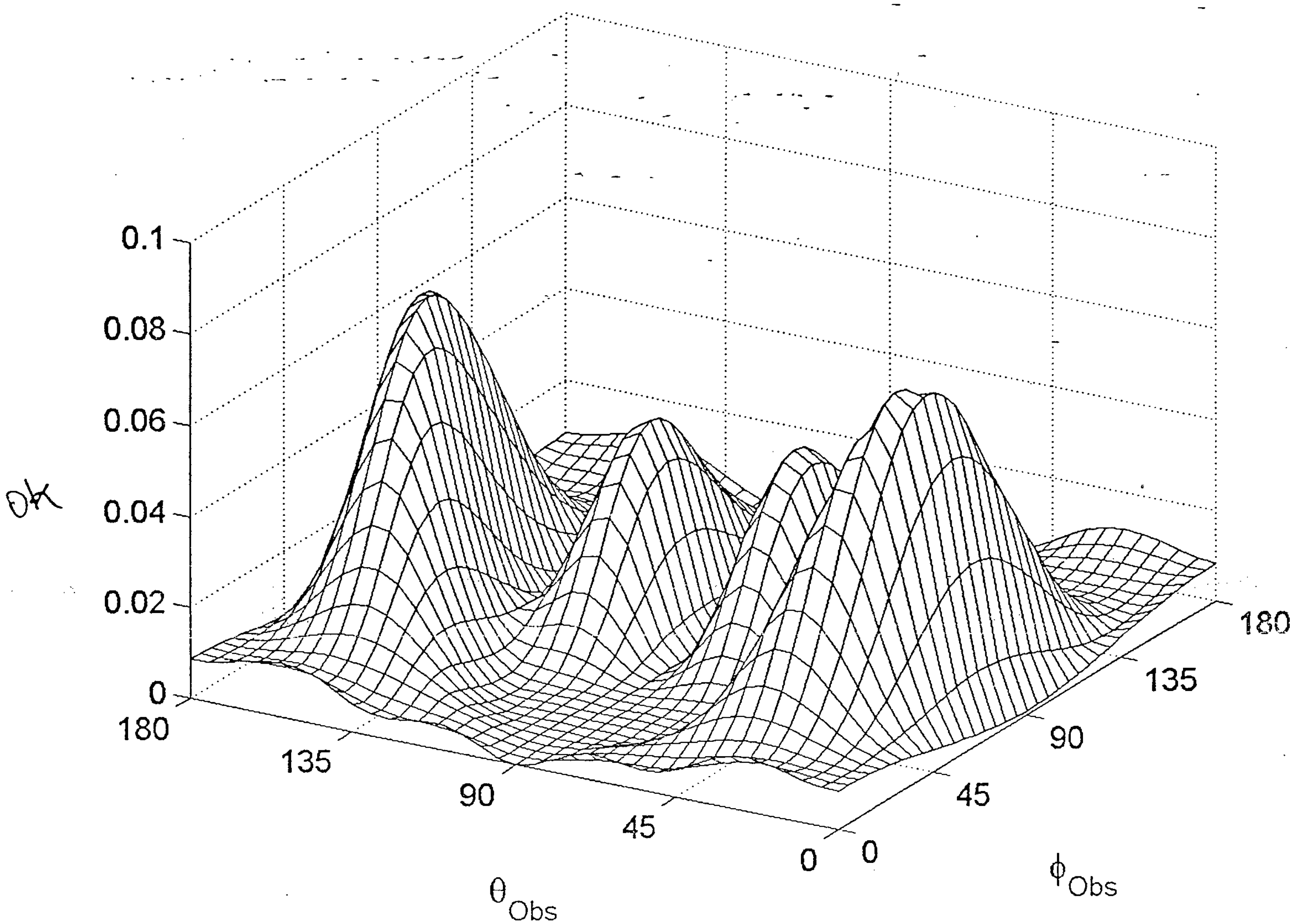
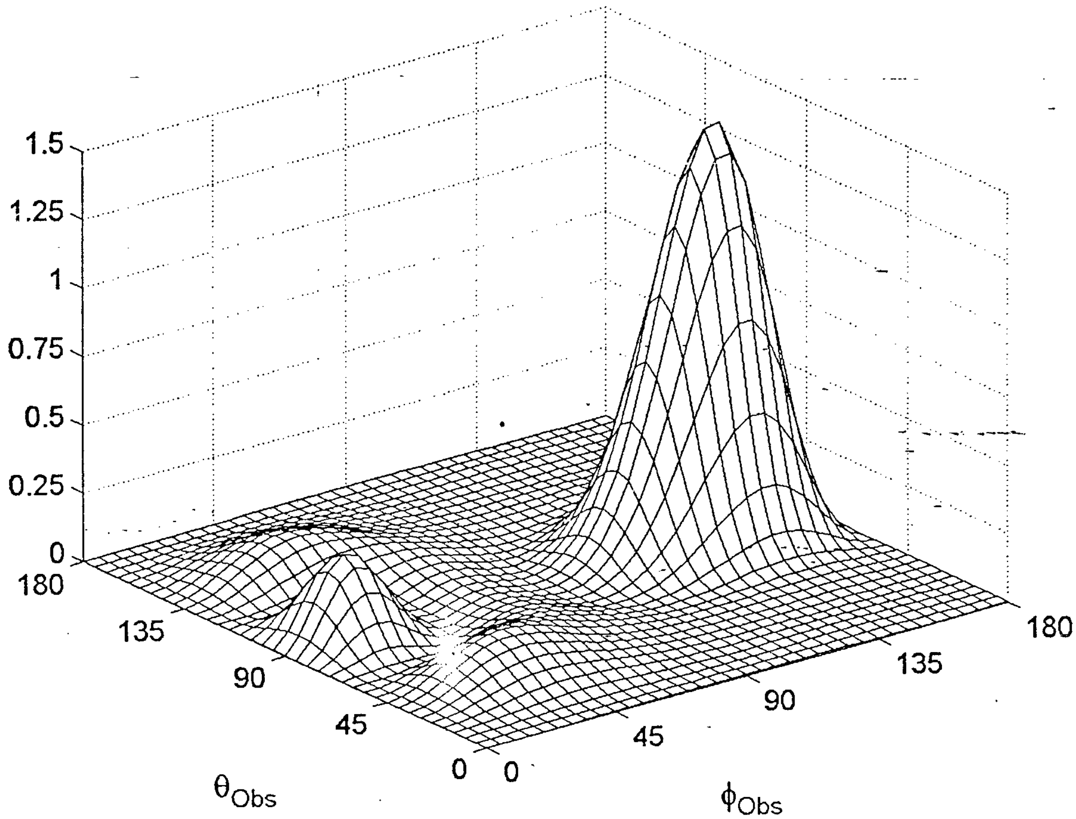
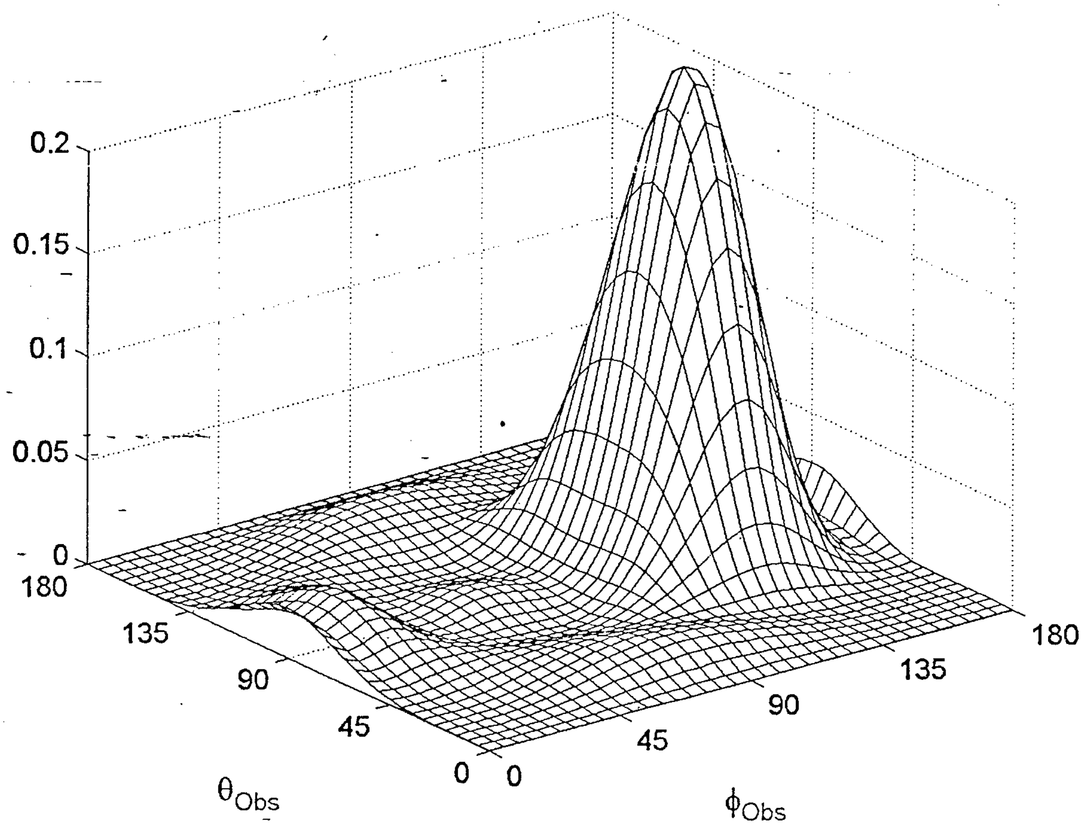


Fig 3d



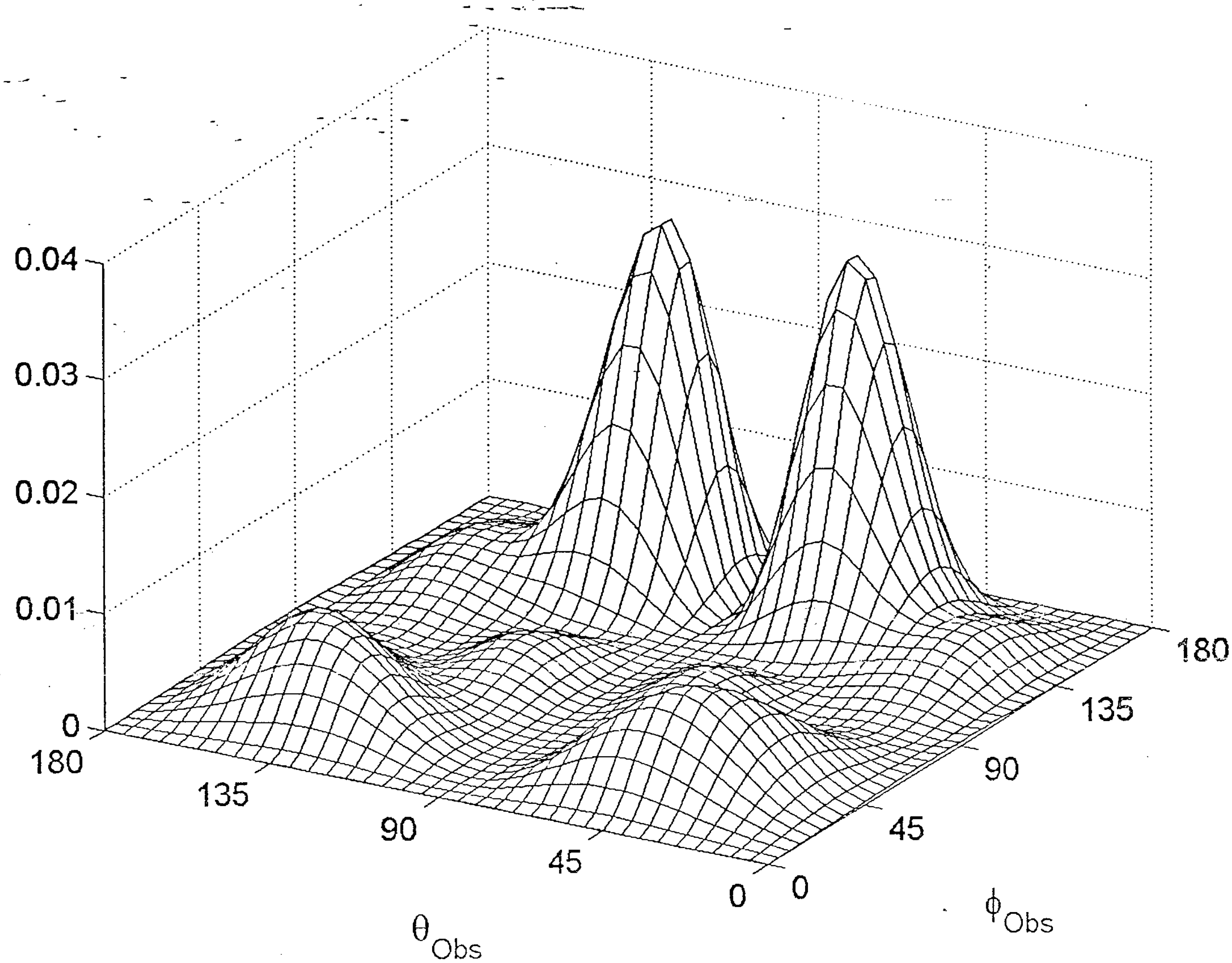
1.5
1.25
1
0.75
0.5
0.25
0
180
135
90
45
0
θ_{Obs}
0
45
90
135
180
ϕ_{Obs}

Fig 4a



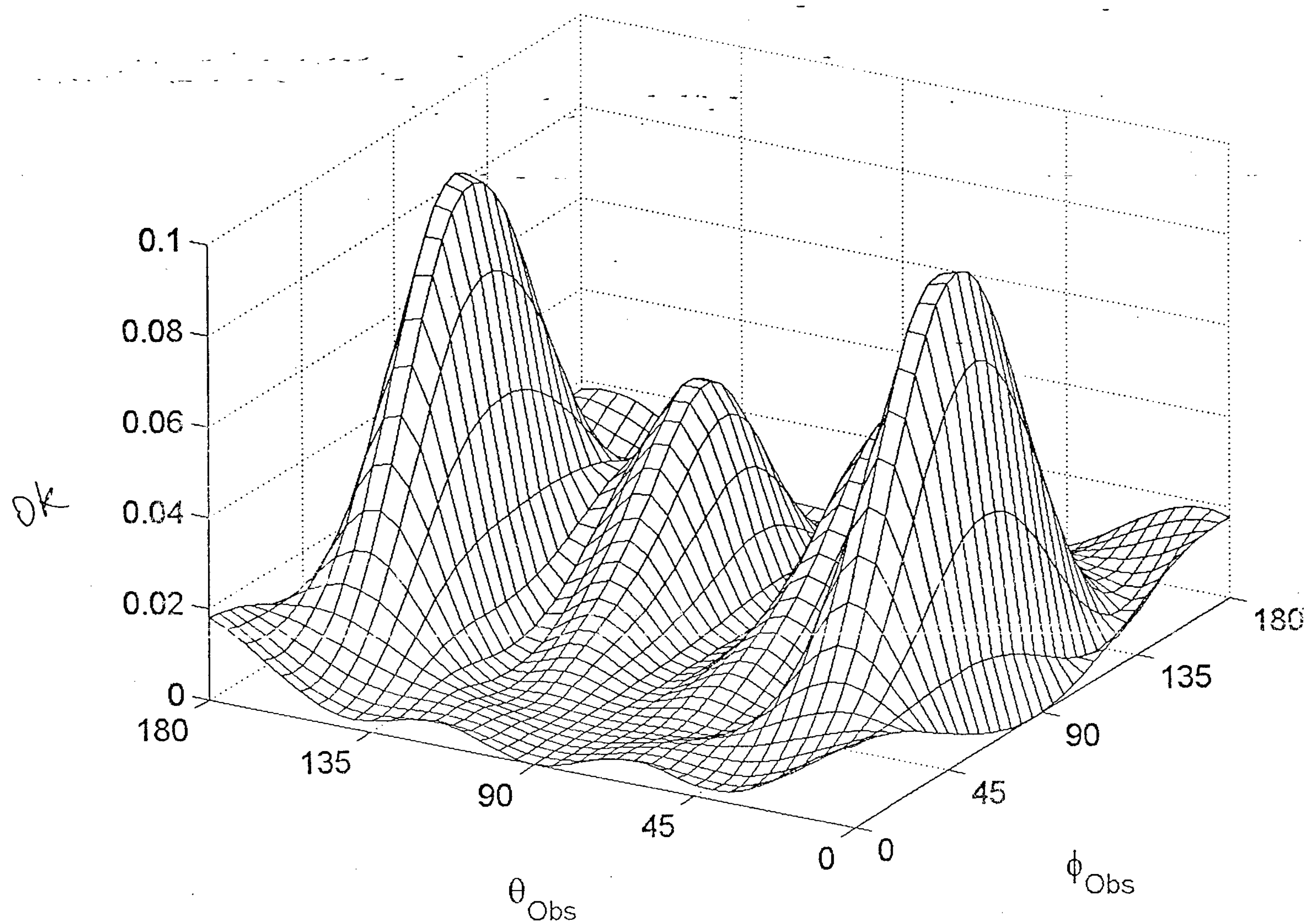
P. Dorle et al. Optical properties...

Fig. 4b



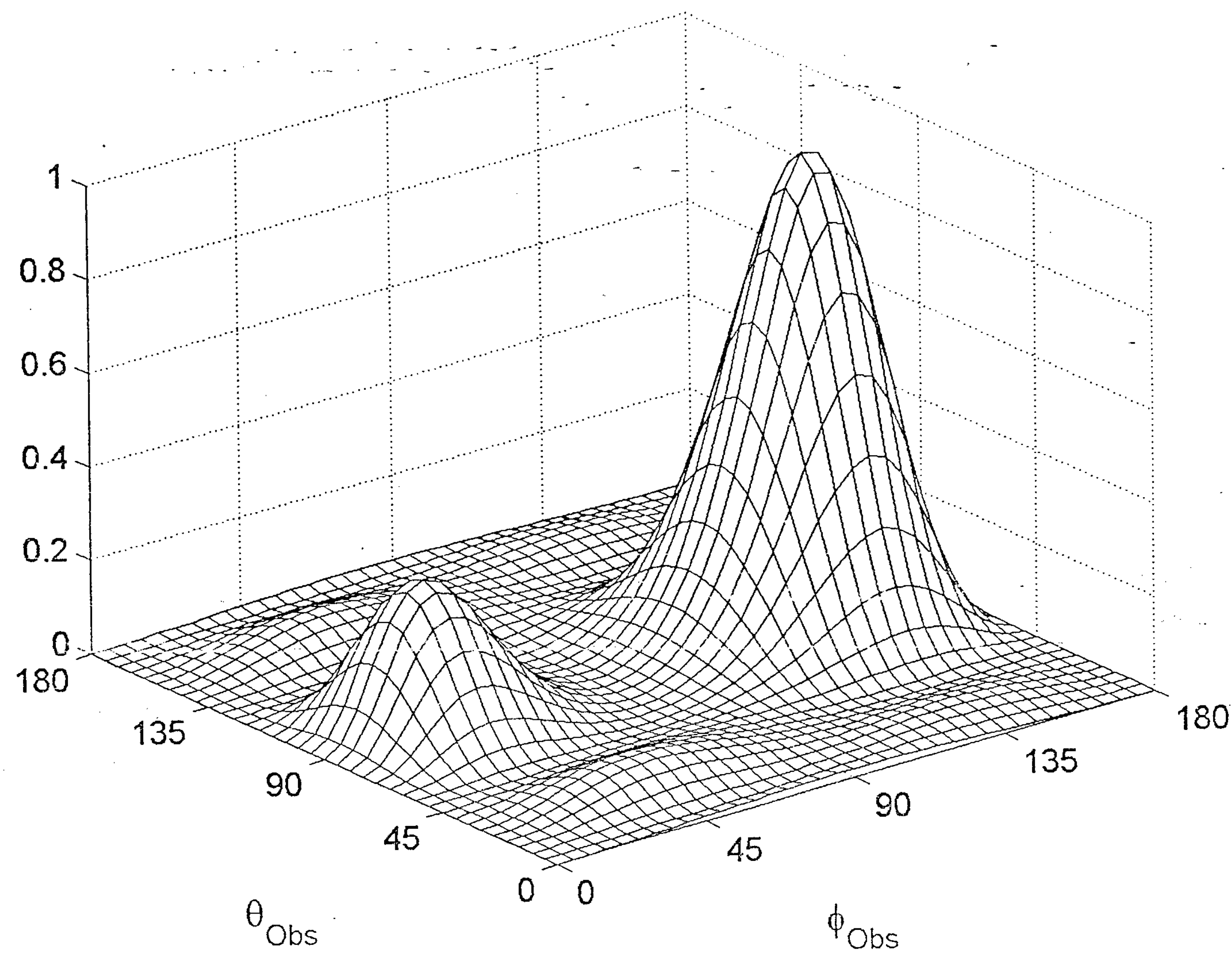
P. Denk et al. Optical Properties...

Fig. 4c



P. Denk et al. Optical properties

Fig. 4d



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